Dynamics of Self-Replicating Patterns in Reaction Diffusion Systems

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Recently solutions to a simple reaction diffusion system have been discovered in which localized structures (spots) make copies of themselves. In this Letter we analyze the one-dimensional analog of this process in which replication occurs until the domain is filled with a periodic array of spots. We provide a heuristic explanation of why this replication process should occur in a broad class of systems. Time dependent solutions are developed for model systems and their analytic structures investigated.

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Over the past three decades the study of self-organization in far-from-equilibrium systems has become a major field of scientific inquiry. Within this field, the study of chemically reacting and diffusing (RD) systems has attained the status of paradigm. Although there are many reasons for the role that RD systems play, perhaps the most compelling is their obvious relevance for biological systems.

Recently, Pearson [1] has observed spot patterns in a RD system that replicate themselves until they occupy the entire domain. This observation was made during a successful attempt to reproduce the labyrinthine patterns observed in [2]. In this Letter, we will look at this model system in one dimension and derive several analytic solutions to the nonlinear partial differential equations, including replicating spot structures. The model [3] is given by

$$\frac{\partial u}{dt} = \nabla^2 u - uv^2 + A(1-u),$$
$$\frac{\partial v}{dt} = \delta^2 \nabla^2 v + uv^2 - Bv. \tag{1}$$

Here $u(x,t)$ and $v(x,t)$ are fields representing the concentrations of two chemical species with differing diffusion coefficients, whose ratio is δ^2. A and B are parameters describing a feed from an external reservoir with the fixed concentrations $u = 1$ and $v = 0$.

One of the most interesting structures observed by Pearson in 2D simulations consists of localized regions of high v and low u concentration, "spots," surrounded by regions where the concentrations are nearer to the kinetics' only fixed point: $u = 1$, $v = 0$. The spots *replicate*: a single spot first divides into two new spots, which then separate until another replication occurs, finally filling the entire domain. Typical asymptotic configurations in these 2D simulations depend on the parameters and consist of either chaotic states in which the spots compete for territory in a continuous process of replication and death or steady states where the spots form a hexagonal pattern.

In 1D, an analogous situation is realized for small enough δ, although the time asymptotic state is always static in a finite domain. Figure 1 is a space-time plot of v for this case. Related phenomena have been observed by other authors [4]. In particular, Kerner and Osipov have derived numerous results on self-organization processes in active media including an analysis of the static division of one-dimensional pulses as the system size is changed.

Recently, replicating spot patterns have been observed experimentally in a RD system [5]. This occurs despite

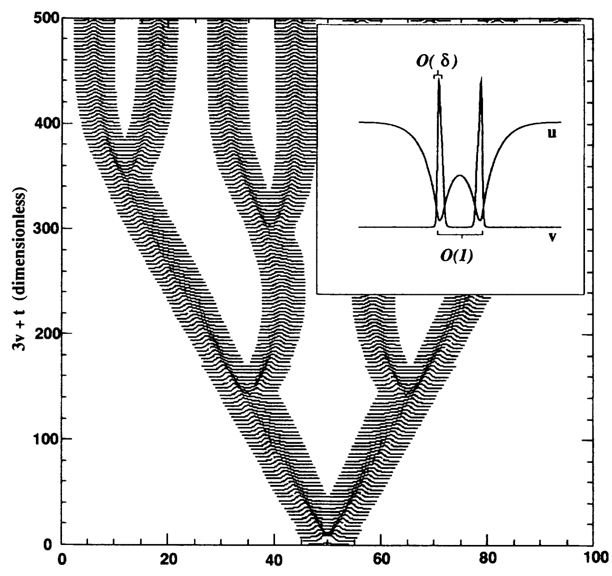

FIG. 1. A space-time plot of v. Regions where v does not vary have been omitted for clarity. Note the repeated replication until a steady state is reached. Inset: A plot of both u and v for two spots. For this simulation, $A = 0.02$, $B = 0.079$, and $\delta^2 = 0.01$. A grid of size 500 was used with a lattice spacing of 0.2.

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the obvious differences in the kinetic details of this system and (1). This fact suggests that replication is very common, characterizing a broad class of RD systems. In this Letter, we present some arguments in support of this proposition, including a heuristic description of the process of replication and demonstrations of analytic features common to several related model RD systems.

One can understand spot formation heuristically. We consider a 1D spatial domain fed with a highly motile, flammable fuel. If this fuel becomes sufficiently hot, the autocatalytic process of burning commences. We further assume that flames spread (diffuse) much slower than the fuel [6]. If a small region is ignited, a local depletion of the fuel occurs. Therefore, gradients are created that induce a lateral flux of fuel into the spot. Thus, the fuel is funneled into the spot from the unignited, "quiescent" regions that surround it.

The fate of an ignited spot depends on the lateral diffusive flux of fuel into it. If this flux is small enough, the system can sustain a steady, localized region of high temperature. As the fuel flux increases, the spot grows. This situation can arise if either the external feed increases, or if the size of the quiescent regions around the spot increases. At some point, the width of the spot will become so large that the flux of fuel into the spot's center will become too low to sustain the autocatalytic reaction there. Under these circumstances, the center of the spot will collapse, creating two spots where before there was one. We refer to this process as "replication." The two spots will now move away from one another as they try to accommodate the fuel flux from the quiescent zones. The region between the spots, which increases in size as the spots move apart, will gradually approach the quiescent state, causing the flux into the back of the two countermoving spots to increase. This increase can induce a secondary splitting in the traveling spots.

We should emphasize that an element crucial for replication is the ability of the spot to autocatalytically consume as much fuel as is provided it, since this depletion is at the heart of splitting. RD models of excitable media, such as the Fitzhugh-Nagumo and Oregonator models [7], do not possess this feature, since their autocatalytic kinetics saturate at some high value of the temperature. This limits the amount of fuel they can consume, and prevents the central collapse characteristic of splitting. The Selkov model of glycolysis [8], which also exhibits replicating spot patterns, is similar to these others in that it does possess a limiting burning branch; however, this branch is located at such a high level of temperature that, over some parameter regime, the entire burning-depletion-collapse cycle occurs well below it.

We can identify each term in (1) with an aspect of our heuristic description. For (1), v plays the role of fire or temperature and u of fuel. The term $A(1-u)$ represents the uniform feed of fuel throughout the domain. The term $-uv^2$ represents the depletion of u as the temperature is increased, $-Bv$ the coupling to the thermal reservoir, and $+uv^2$ the autocatalytic production of heat. For values of v below the autocatalytic branch ($uv - B = 0$) the kinetics relaxes back to $u = 1$, $v = 0$. For v above this branch, autocatalytic "burning" commences [6] and as u is depleted, eventually v will be pulled back below the branch, returning to its steady value.

These qualitative arguments do not depend on a specific set of kinetics. If we replace the term $-uv^2$ in the first of (1) with $-v$, we obtain the so-called "linear model." The new term still reflects the depletion of fuel with increasing temperature and this model also exhibits the phenomenon of replication. For the linear model, the singular perturbation analysis we develop in this Letter carries over directly; however, for other models, e.g., the Selkov model, this analysis does not apply.

We now turn to analytic solutions of the system (1) that correspond to the evolution observed in Fig. 1. We can construct these solutions in the singular limit $\delta \to 0$. The simulations show that the spatial domain is divided into "inner" or spot regions of width $\mathcal{O}(\delta)$ and "outer" regions of width $\mathcal{O}(1)$ (see Fig. 1). In the inner regions $v \sim \delta^{-1}$ and $u \sim \delta$, while in the outer regions, v is transcendentally small and u is of order 1. There are two characteristic time scales. The spots evolve slowly over long time intervals that are punctuated by replication which occurs on a fast time scale. The solutions which we construct are valid only during the long slow intervals but they do predict when and why replication occurs.

In both the inner and outer regions we introduce the slow time $\tau = \delta\sqrt{B}t$. The following rescaled variables are appropriate in the inner regions: $u_{\rm in} = \delta\sqrt{B})^{-1}u$, $v_{\rm in} = \frac{\delta}{\sqrt{B}}v$, and $x_{\rm in} = \frac{\sqrt{B}}{\delta}[x - x^m(\tau)]$, where x^m is the location of the minimum of u in the spot under consideration. We define $c(\tau) \equiv dx^m/d\tau$ as the rescaled velocity. In the outer regions we set $v = 0$, $u = u_{\rm out}$ and do not rescale x. With these scalings, the evolution equations (1) reduce to

$$\frac{\partial^2 u_{\rm out}}{\partial x^2} + A(1 - u_{\rm out}) = \mathcal{O}(\delta) \tag{2}$$

in the outer regions and

$$\frac{\partial^2 u_{\rm in}}{\partial x_{\rm in}^2} - u_{\rm in}v_{\rm in}^2 = \mathcal{O}(\delta),$$

$$\frac{\partial^2 v_{\rm in}}{\partial x_{\rm in}^2} + c(\tau)\frac{\partial v_{\rm in}}{\partial x_{\rm in}} + u_{\rm in}v_{\rm in}^2 - v_{\rm in} = \mathcal{O}(\delta) \tag{3}$$

in the inner regions, which we solve as power series in δ [9]. Note that to leading order in δ, the feed of u drops out of (3). This is consistent with the spots being maintained solely by the *lateral* diffusive flux of *fuel* from the surrounding outer regions. Also note that with this scaling $\partial^2 u_{\rm in}/\partial x_{\rm in}^2 \geq 0$. Thus, it is clear that we cannot describe pulse division of the $u_{\rm in}$ field. However, replication is always preceded by the $v_{\rm in}$ field developing a dimple and this feature is retained with the current scaling. The fact that the explicit time dependence scales out of the system in both the inner and outer regions means that the variation with τ is adiabatic; thus the time dependent quantities in the problem are treated as constants.

We obtain a single spot solution to Eq. (1) in the domain $[x_-^M, x_+^M]$, where x_-^M and x_+^M are the locations of two successive maxima of u. The resulting solution can be used to construct an array of moving spots on a larger (possibly infinite) domain. Static solutions correspond to the particular case $c = 0$. Since u has only one minimum in $[x_-^M, x_+^M]$, the interval is divided into three regions: the outer regions to the right and left of the spot, and the inner region centered at $x^m(\tau)$.

The solutions in the different regions must go smoothly into one another. Thus the $\mathcal{O}(1)$ terms in the left and right outer solutions, u_{out}^- and u_{out}^+, must be zero where the outer regions meet the inner, i.e., at $x \approx x^m$. Thus the solutions in the left and right outer regions can be written

$$u_{\text{out}}^{\pm}(x,\tau) = 1 - \hat{u}^{\pm} \cosh[\sqrt{A}(x - x_{\pm}^M)], \tag{4}$$

where $\hat{u}^{\pm} = \operatorname{sech}[\sqrt{A}(x^m - x_{\pm}^M)]$.

As we approach the spot, we find from these equations that $u_{\text{out}}^{\pm} \approx \pm L_{\text{out}}^{\pm}(x - x^m)$, where

$$L_{\text{out}}^{\pm} \equiv \sqrt{A} \tanh(\sqrt{A}|x_{\pm}^M - x^m|) \tag{5}$$

are the diffusive fluxes of u into the spot. Note that if $x_{\pm}^M \to \pm\infty$, then $L_{\text{out}}^{\pm} = \sqrt{A}$.

We determine solutions to Eqs. (3) that are consistent with the outer ones as $x_{\text{in}} \to \pm\infty$. This implies the behaviors $u_{\text{in}} \sim \pm L_{\text{in}}^{\pm} x_{\text{in}} + M^{\pm}$, $v_{\text{in}} \sim v_{\infty}^{\pm} \exp(\frac{-c \mp \sqrt{c^2+4}}{2} x_{\text{in}})$ as $x_{\text{in}} \to \pm\infty$. These behaviors are consistent with (4) in the limit $x - x^m \to 0^{\pm}$. By unscaling the inner variables, we find that $u \cong u_{\text{in}} \delta \sqrt{B} \sim \pm B L_{\text{in}}^{\pm}(x - x_m) + \mathcal{O}(\delta)$ and $v \cong \frac{v_{\text{in}}\sqrt{B}}{\delta}$ is a transcendentally small quantity as $x_{\text{in}} \to \pm\infty$. Matching gives the relation $L_{\text{in}}^{\pm} = B L_{\text{out}}^{\pm}$.

For a given pair of fluxes, L_{in}^+, L_{in}^-, there are five unknowns characterizing the solutions: $v_{\infty}^{\pm}$, $M^{\pm}$, and c. Using the asymptotic behaviors as initial conditions, we integrate Eqs. (3) to $x_{\text{in}} = 0$ from $x_{\text{in}} \sim \pm\infty$. We require continuity of u_{in}, v_{in} and their derivatives, and impose $\frac{\partial u_{\text{in}}}{\partial x_{\text{in}}}|_{x_{\text{in}}=0} = 0$, which breaks the degeneracy due to the translational invariance of (3). This gives five constraints on the five unknowns as functions of the fluxes L_{in}^+ and L_{in}^-. In general, we expect a discrete set of solutions for given values of $L_{\text{in}}^{\pm}$. This is consistent with our heuristic picture: once we know the amount of fuel that it is being fed from its sides, the structure of the spot can be determined.

We plot the velocity in Fig. 2. Although many solutions were found numerically, corresponding to multi-bumped $v_{\text{in}}(x_{\text{in}})$, we only consider those observed in simulation, the single and double bumped solutions. The surface, $c(L_{\text{in}}^+, L_{\text{in}}^-)$, has two sheets that intersect along the line $L_{\text{in}}^+ = L_{\text{in}}^- < L_c$, where L_c is some critical value. Along this line, the two solutions consist of either symmetric single or double bumped spots, both with $c = 0$. The single bumped solutions are linearly stable and the double bumped unstable. These two branches collide and disappear at $L_{\text{in}} = L_c$. We have also observed this disappearance in the full system (1). For small enough δ we have continued its static solutions finding the same value L_c as predicted by the perturbation theory. These static solutions are identical to those discussed by Kerner and Osipov [4]. If one starts on one of the static inner solutions and continues it in the L_{in}^+, L_{in}^- plane, around the singularity at L_c one arrives at the second static solution. Hence, the two sheets represent single and double bumped solutions which go smoothly into one another. The solutions disappear as L_{in} is increased. For this case, it appears that the c sheets in the L_{in}^+, L_{in}^- plane are colliding with another sheet of solutions, although our numerics cannot confirm this. This disappearance corresponds to replication.

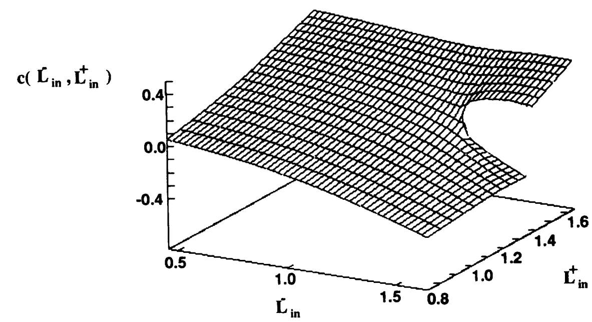

FIG. 2. A plot of the function $c(L^+, L^-)$. All axes have dimensionless units.

Given that $c = \frac{dx^m}{d\tau}$, the function $c(L_{\text{in}}^+, L_{\text{in}}^-)$ together with the relations (5) determine the evolution of our spot solution. If we solve for an array of N moving spots, located at x_i^m, then the locations of the maxima of u, x_i^M, $0 < i < N$ are functions of τ related to their adjacent minima by $x_i^M = (x_{i+1}^m + x_i^m)/2$. The dynamics of the x_i^m's are determined by

$$\frac{dx_i^m}{d\tau} = c(L_i^+, L_i^-), \tag{6}$$

where the fluxes $L_i^{\pm}$, $1 \leq i < N$ are functions of $x_{i+1}^m - x_i^m$ and $x_i^m - x_{i-1}^m$, via a generalization of Eq. (5). The fluxes at the ends of the domains are determined from the boundary conditions. Equations (6) can be integrated forward in time while all the functions $c(L_i^+, L_i^-)$ exist for the given set of fluxes. If at some time $c(L_i^+, L_i^-)$ no longer exists for a spot, then that spot replicates. After this occurs, the new configuration can be used as an initial condition to integrate Eqs. (6) up to the next splitting.

We have compared the solutions obtained in this way with the numerical simulations of the full system (1), finding good agreement in the shapes of the pulses and the value of the velocity. We have also verified that the spot's splitting occurs when the fluxes into it are such that the inner solutions disappear. Finally, we have calculated the linear stability of these solutions. We find that many of the traveling solutions that are observed in simulation (in particular those that are developing a second bump in the v field) are linearly unstable. We conclude that nonlinear effects must be restabilizing the spots, which persists until their dynamics actually carry them to a point where the solution ceases to exist—at which point replication occurs.

Is it reasonable to expect that the structure of the function $c(L_{\rm in}^+, L_{\rm in}^-)$ should also be characteristic of a broad class of models? To check this proposition, we have applied the same analysis to the linear model, where we find the same type of structure for the selected velocity. We believe that the double sheeted nature of the surface $c(L_{\rm in}^+, L_{\rm in}^-)$ is in some sense a "fingerprint" of replication, and should be amenable to more general analytical approaches. In particular, we surmise that the analytic structure of the singularity at $L_{\rm in}^+ = L_{\rm in}^- = L_c$ is characteristic of all of these structures. We do not yet have an analytic formulation for the solutions in the neighborhood of this singularity. Verification of this proposition is a subject of future research.

From the physical point of view, the analysis presented here is crude. In most experiments the ratio of diffusion coefficients is much closer to unity. However, the singular limit presented here clarifies which physical processes are dominant as the system evolves. An analysis for δ close to but greater than 1 would be of interest. In higher dimensions the necessary analysis is far more delicate as can be seen from the fact that in our analysis the parameter B scales out of the problem and in [1] the solutions depend strongly on the values of A and B. The role that curvature plays is clearly nontrivial since the coupling between the spot and the outer regions is curvature dependent.

In closing we comment that the fuel and fire picture also provides an understanding of the Turing patterns which bifurcate off of the uniformly burning branch. Indeed it is possible to view spot replication as the dynamical mechanism by which Turing patterns spread for parameter values well beyond onset. Near onset, hexagonal patterns that nucleate locally spread by spot replication [10] until the domain is filled with small-amplitude static hexagons. The fuel and fire picture is equivalent to the standard activator-inhibitor picture which was developed to provide an intuitive explanation to elucidate the mechanism of Turing instability. The fuel is, perhaps paradoxically, the *inhibitor* and temperature the *activator.*

In order to simplify the analysis of Turing instabilities, skeletal models were created in which the only steady state of the reaction kinetics is a *nonequilibrium* steady state. The Brusselator [11] is the premier example of such a model. Clearly, one could relax the set of assumptions used in obtaining such models so as to keep the thermodynamic branch. We expect that most extended versions of the Brusselator [of which Eq. (1) is an example] that keep the thermodynamic branch would exhibit the phenomena discussed here.

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[1] J. E. Pearson, Science **261**, 189 (1993).
[2] K. J. Lee, W. D. McCormick, Q. Ouyang, and H. L. Swinney, Science **261**, 192 (1993).
[3] P. Gray and S. Scott, Chem. Eng. Sci. **38**, 29 (1983).
[4] T. C. Lacalli, Philos. Trans. R. Soc. London **294**, 547–588 (1981); A. B. Rovinsky, in *Nonlinear Wave Processes in Excitable Media*, edited by A. V. Holden *et al.* (Plenum, New York, 1989); P. Arcuri and J. D. Murray, J. Math. Biol. **24**, 141 (1986); J. Gorski and A. L. Kawczynski, Polish J. Chem. **58**, 61 (1985); B. S. Kerner and V. V. Osipov, Usp. Fiz. Nauk. **160**, 1 (1990) [Sov. Phys. Usp. **33**, 3 (1990)].
[5] K. Lee, J. E. Pearson, W. D. McCormick, and H. L. Swinney (to be published).
[6] We are not describing actual combustion; rather we want to provide a *qualitative* explanation of the phenomenon of self-replication in *chemical* reaction diffusion systems. Thus, we ignore radiative and convective effects.
[7] D. A. Kessler, H. Levine, and W. N. Reynolds, Physica (Amsterdam) **70D**, 115 (1994).
[8] E. Selkov, Eur. J. Biochem. **4**, 79 (1968).
[9] For brevity, we will only keep terms of $\mathcal{O}(1)$ in our expansions. The full theory does, of course, treat these higher order corrections but the salient behavior is captured by the truncated equations. Details of the higher order terms and their matching between the two regions can be found in [12].
[10] P. de Kepper, J. J. Perraud, D. Rudovics, and E. Dulos (to be published).
[11] R. Lefever, G. Nicolis, and I. Prigogine, J. Chem. Phys. **47**, 1045 (1967).
[12] W. N. Reynolds, S. P. Dawson, and J. E. Pearson (to be published).